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Filonenko, K.; Willatzen, Morten; Bordo, V.

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Spontaneous Emission Enhancement at Finite-length Metal Nanowires

K. Filonenko[1], **M. Willatzen**[2], **and V. Bordo**[1]

[1]NanoSyd, Mads Clausen Institute, Syddansk Universitet, Alsion 2, DK-6400 Sønderborg, Denmark
[2]Department of Photonics Engineering, Technical University of Denmark
DTU Fotonik, Building 345 West, DK-2800 Kongens Lyngby, Denmark

Abstract— We study spontaneous emission enhancement of a two-level atomic emitter placed in a dielectric medium near a finite-length cylindrical metal nanowire. We calculate the dependence of the Purcell factor and the normalized decay rate to a continuous spectrum on the nanowire radius for several emitter transition wavelengths and different orientations of the transition dipole moment. For a particular transition wavelength we calculate the dependence of these quantities as well as the β-factor on the emitter distance from the nanowire and the nanowire radius. The obtained results demonstrate that the spontaneous emission characteristics exhibit significant differences as compared to the case of an infinite wire.

1. INTRODUCTION

Recently, growing attention has been focused on control over spontaneous emission of quantum emitters coupled to metal nanowires [1–10]. Such a system is not only of fundamental importance but also promises various applications, including single-plasmon sources [3], single-photon transistors [11], plasmonic amplifiers [12] and resonators [10], and nanowire-based spasers [13].

The conventional theoretical model which is used to calculate the emitter decay rate assumes that a metal nanowire has a cylindrical shape and is infinite in its axial dimension. Such an approach does not allow to describe the reflections from the nanowire end facets and hence the modes of the plasmonic nanocavity. In this paper, we overcome this drawback by applying a rigorous approach developed in Refs. [15, 16] which gives the dyadic Green's function for a cylindrical nanocavity. We demonstrate that taking into account finiteness of the nanowire length leads to considerably different results for the Purcell and β-factors as compared to the infinite-length approximation.

2. DEFINITIONS

The total spontaneous emission rate of an emitter in the presence of a cavity can be represented as a sum of different contributions:

$$\gamma^{\mathrm{tot}} = \sum_k \gamma^k + \gamma^{\mathrm{cs}}, \tag{1}$$

where γ^k is the decay rate to k-th discrete mode of the cavity and γ^{cs} is the decay rate to a continuous spectrum of modes. The corresponding normalized decay rates are then connected through

$$\frac{\gamma^{\mathrm{tot}}}{\gamma^0} = \sum_k F_p^k + \frac{\gamma^{\mathrm{cs}}}{\gamma^0}, \tag{2}$$

where γ^0 denotes the decay rate in an unbounded medium and

$$F_p^k = \frac{\gamma^k}{\gamma^0} \tag{3}$$

is the Purcell factor for the k-th discrete mode. The ratio

$$F_\beta^k = \frac{\gamma^k}{\gamma^{\mathrm{tot}}} = \frac{F_p^k}{\gamma^{\mathrm{tot}}/\gamma^0} \tag{4}$$

is called the β-factor and is extensively used in cavity quantum electrodynamics.

In this paper, we calculate the Purcell and β-factors for a dipole emitter located in the vicinity of a silver nanowire associated with the nanowire fundamental mode TM0. We compare the contributions of both discrete and continuous spectrum modes to the total normalized rate (2) for different orientations of the emitter dipole moment.

3. THEORETICAL MODEL

The nanowire is modeled by a silver cylinder of radius a, length L, and a wavelength-dependent dielectric permittivity $\epsilon_2(\lambda)$, which is found as an interpolation of the data for complex refractive index given in [17]. We use the approximation of an elongated nanowire [15], assuming that the length of the wire is much greater than its radius: $L \gg a$. A two-level quantum emitter with transition frequency ω, the corresponding wavelength $\lambda = (2\pi c)/\omega$ and the dipole momentum $\vec{\mu}$ is placed in the outer medium with constant permittivity $\epsilon_1 = 2$ at a distance d from the wire as is shown in Figure 1(a). We consider the radii in the range 10–50 nm and the emitter transition wavelengths in the range 500–1250 nm. In these ranges a silver nanowire supports two surface plasmon polariton modes: a transverse magnetic mode TM0 and a hybrid mode HE1. All other modes are cut off at higher radius values and therefore do not contribute to the total spontaneous emission rate in the considered range. Although the HE1 mode does not have a cutoff radius as all other higher order modes, it was shown in [2] that its mode volume grows exponentially as the wire radius gets smaller and therefore the mode is effectively cut off at very small radius values considered here. We therefore limit ourselves to a single-mode regime and calculate the dependence of the longitudinal wavenumber β on the radius a (Figure 1(b)) for several transition wavelengths ($\lambda = 500$ nm, 750 nm, 1000 nm and 1250 nm).

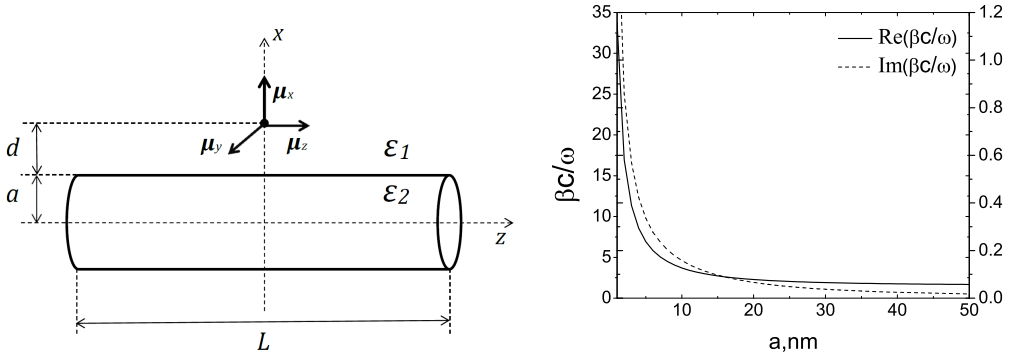

Figure 1: The model of the system: (a) Scheme showing the metallic wire and the orientations of the emitter along the axes of the rectangular coordinate system μ_x, μ_y, μ_z, (b) TM0 mode dispersion: real part of the normalized longitudinal wavenumber (left axis) and its imaginary part (right axis) as a function of the nanowire radius.

The electromagnetic field is described in terms of the electric and magnetic Hertz potentials, $\mathbf{\Pi}^e$ and $\mathbf{\Pi}^m$, satisfying the Helmholtz equations inside ($j = 2$) and outside ($j = 1$) the cylinder:

$$\nabla^2\mathbf{\Pi}^e(\mathbf{R}) + k_j^2\mathbf{\Pi}^e(\mathbf{R}) = -\frac{4\pi}{\epsilon_j}\mathbf{P}_j(\mathbf{R}), \tag{5}$$

$$\nabla^2\mathbf{\Pi}^m(\mathbf{R}) + k_j^2\mathbf{\Pi}^m(\mathbf{R}) = 0, \tag{6}$$

where $\mathbf{P}_j = \vec{\mu}\delta_{1j}\delta(\mathbf{R} - \mathbf{R}_0)$ is the electric polarization density of the external source, δ_{ij} is a Kronecker delta, $\delta(\mathbf{R} - \mathbf{R}_0)$ is Dirac's delta function centered at the position of the emitter, $\mathbf{R}_0$, and $k_j = (2\pi/\lambda)\sqrt{\epsilon_j(\lambda)}$ is the modulus of the wave vector in the medium with index $j \in \{1, 2\}$. The solutions to these equations dictate the electric and magnetic fields in both media [18]. In the next section, we calculate and plot different contributions to the normalized decay rate (2) and the β-factor using the relation [19]

$$\gamma^{\text{tot}} = \frac{2}{\hbar}\sum_{\alpha\beta}\mu_\alpha\mu_\beta \text{Im}\bar{\mathbf{G}}_{\alpha\beta}(\mathbf{R}, \mathbf{R}_0; \beta(a)), \tag{7}$$

where $\bar{\mathbf{G}}_{\alpha\beta}(\mathbf{R}, \mathbf{R}_0; \beta(a))$ are the rectangular components of the dyadic Green function of the system which was derived in [16] to describe the finite-length cylinder geometry.

4. RESULTS AND DISCUSSION

The rigorous analysis based on the theory developed in [16] allows one to make the following conclusions about the spontaneous emission to the TM0 mode in the limit $L \gg a$: (1) the Purcell

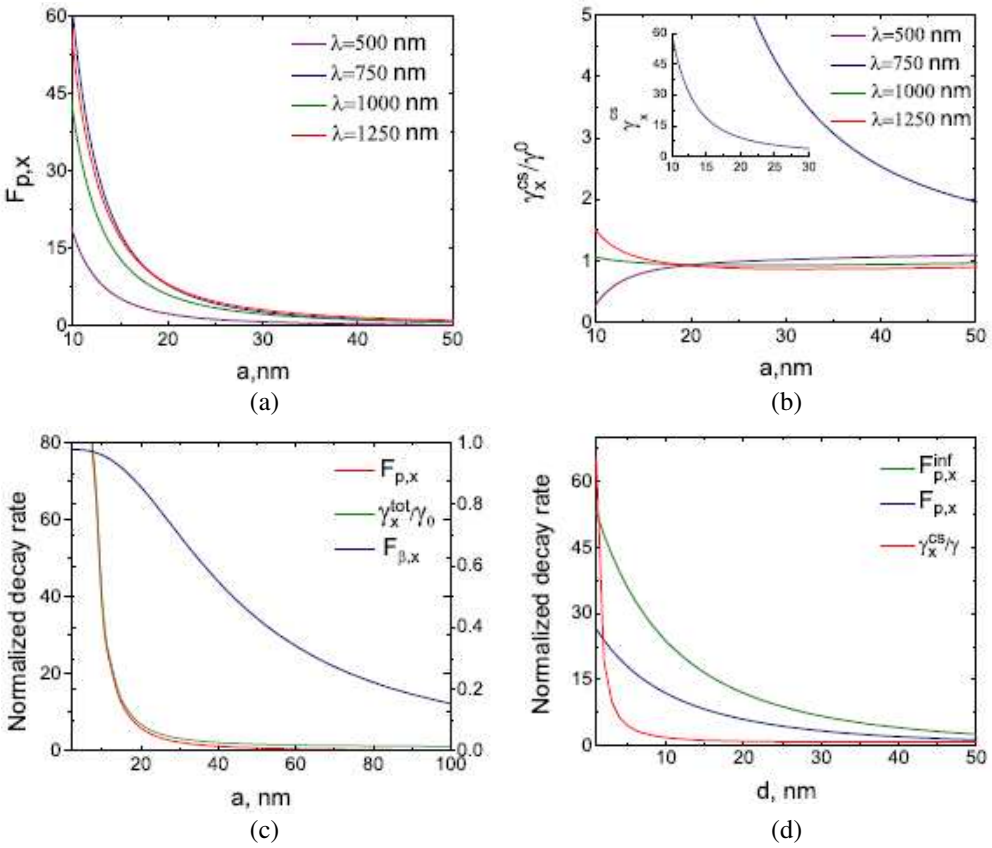

Figure 2: Normalized decay rates for a x-oriented dipole emitter: (a) the Purcell factor and (b) the normalized decay rate to a continuous spectrum plotted as a function of the nanowire radius for several emitter wavelengths λ with the emitter-wire distance taken as $d = a$, (c) the Purcell factor (red curve, left axis), the decay rate to a continuous spectrum (green curve, left axis), and the β-factor (blue curve, right axis) as a function of the radius a with $d = a$, $\lambda = 1\,\mu\text{m}$ and $\epsilon_2 = -50 + 8.5i$, (d) the Purcell factor in the limit of an infinite nanowire (green curve), the Purcell factor in the limit of an elongated nanowire (blue curve) and the decay rate to a continuous spectrum (red curve) as a function of the emitter-wire distance d with $a = 25\,\text{nm}$, $\lambda = 1\,\mu\text{m}$, $\epsilon_2 = -50 + 8.5i$.

factor for this mode does not depend on the length of the wire and the position of the emitter along the z-axis; (2) the mode is not excited if the dipole moment of the emitter is oriented along the y-axis. The first of these facts is in agreement with the statement, proved in [20, 21] for a semi-infinite wire, that the far-field reflection coefficient from the end facets for such a mode is equal to zero. As a consequence the mode does not have a Fabry-Perot-like structure.

We plot the normalized decay rates for the x-orientation of the emitter transition dipole moment $\vec{\mu}$ in Figure 2 as a function of the wire radius and the wire-emitter distance. Analogous results for z-orientation are similar and are not shown here. These plots give rather high Purcell factor values signifying effective excitation of the surface plasmon polariton mode. The dependence of the Purcell factor on the nanowire radius is calculated for several transition wavelengths and is represented in Figures 2(a) and 2(b). The distance changes along with radius as $d = a$. These figures show that in some range of parameters the decay to the TM0 mode dominates over the other decay channels. This takes place for all radii below a certain value which for $\lambda = 500\,\text{nm}$, 750 nm, 1000 nm, and 1250 nm is equal to 27 nm, 11 nm, 45 nm, and 53 nm, respectively, for the x-orientation of the transition dipole moment and to 37 nm, 50 nm, 33 nm, and 31 nm for the z-orientation. For larger radii the other decay channels start to dominate. For example, in the case of the x-orientation and for $\lambda = 750\,\text{nm}$ the normalized decay rate to the continuous spectrum is greater than the Purcell factor over the whole relevant range starting from 11 nm (blue curve in Figures 2(a) and 2(b)). This can be changed, however, by placing an emitter closer to the wire because, as is seen from Figure 2(d), the Purcell factor increases with decreasing distance.

In Figure 2(c) the Purcell factor (red curve) is compared with the total decay rate (2) (green

curve) for $\lambda = 1\,\mu$m depending on the nanowire radius. These quantities appear to be very close to each other for small radius values. Correspondingly, the β-factor plotted in the same figures (blue curve) approaches unity as the wire radius gets smaller. This makes the TM0 mode suitable for utilizing in a nanowire-based spaser. In Figure 2(d) we compare the Purcell factor calculated in the approximation of elongated nanowire (blue curve) with the one calculated in Ref. [7] in the limit of an infinite nanowire (green curve). It is found that these quantities differ by a factor of two in the whole range of the wire-emitter distance, d. This means that the infinite wire approximation largely overestimates the Purcell factor and is not at all accurate, at least for the problem under consideration.

5. CONCLUSION

The rigorous approach developed in Ref. [16] has enabled us to describe the spontaneous emission enhancement for a two-level quantum emitter in the vicinity of a finite-length cylindrical metal nanowire. We have calculated the Purcell and β-factors as well as the decay rate to the continuous spectrum of modes and plotted them as functions of the nanowire radius and the emitter-nanowire distance in the regime where a decay to a single surface plasmon polariton mode is essential. The Purcell and β-factors were found to be simultaneously maximal at extremely small radii and emitter-nanowire distances (below 30 nm which is much smaller than the half of the emission wavelength 500–1250 nm). This makes metal nanowires interesting for nanoscale spaser applications. The obtained results have been compared with the calculations in the approximation of an infinite nanowire. It is found that the infinite nanowire model largely overestimates the Purcell factor magnitude.

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